

# Narrowband multilayer coatings for the extreme ultraviolet range of 50-92 nm

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**Abstract:** A new type of multilayer coatings with narrowband reflection properties and peaked in the ~50–92 nm spectral range has been developed. Multilayers are based on Yb, Al, and SiO films and they have been prepared by thermal evaporation. Efficient multilayers based on Yb and Al, with an SiO protective layer were prepared, but they developed a dendrite structure, which was attributed to the reactivity between Al and Yb. Multilayers based on Yb and Al, with both SiO protective and barrier layers, resulted in efficient reflective filters, with no observable dendrite growth. The peak reflectance of aged multilayers was of the order of ~0.20, with bandwidths in the range of 12 to 22 nm FWHM.

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## 1. Introduction

Coatings with narrowband reflection properties have become available in most spectral ranges. Even in the extreme ultraviolet (EUV), where materials usually absorb radiation much more than they do at visible wavelengths, the use of multilayers has become possible due to the availability of low-absorption materials. At wavelengths slightly longer than 115 nm, narrowband multilayers have been prepared based on  $\text{MgF}_2$  with a second material that was selected among either Al or some fluorides [1–3]. Just below 50 nm, efficient multilayers based on Sc/Si [4–6], and Mg/SiC [7,8] have been developed; they provide useful bands centered in the ~30–50 nm range. However, the ~50–115-nm spectral range has been lacking such coatings because the overwhelming majority of materials is burdened with a strong absorption; this turns inefficient the accumulation of layers in a multilayer.

The number of narrowband reflectance multilayers developed so far within the ~50 – 115 nm spectral range is very limited. Narrowband multilayers based on Tb [9–11], Gd [12,13], Nd [12], and La [11], combined with Si, SiC or  $\text{B}_4\text{C}$ , were developed recently. These multilayers demonstrated useful bands centered in the ~55–69 nm range with peak reflectances in the 0.12–0.27 range. At wavelengths longer than ~69 nm, a multilayer based on La and  $\text{B}_4\text{C}$ , with a peak reflectance of ~0.118 at 92 nm has been reported [11]. The above are the first narrowband multilayers available in this range and it can be expected that multilayers more efficient, and peaked in a wider range, are possible. Other than this, a remarkable transmission filter based on a cold LiF substrate coated with an In film was developed by Carruthers [14].

Besides the aforementioned narrowband coatings, additional multilayers have been proposed to enhance the reflectance at wavelengths that lie in the 60–115 nm [15–21]. However, these multilayers either reflect in wide bands or were designed to provide high reflectance at a specific wavelength and suppression at another one, so that the latter multilayers do not have strictly narrowband performance.

A new type of multilayers peaked in the core of the aforementioned 50 – 115 nm interval is reported here [22]. The multilayers are based on the relative transparency of Yb and on the use of Al as a contrasting material; SiO is the third material of the multilayer and it is used both as a capping as well as a barrier layer. We designed multilayers with peak reflectance at wavelengths within ~50 to 92 nm. The experimental results reported in this work correspond to coatings peaked in the 78 – 91 nm range. These multilayers provide valuable filtering performance in a spectral range where few developments have been reported. Section 2 focuses on multilayer design. The experimental setup and procedures are described in section 3. Section 4 reports on the reflectance measured for multilayers with and without barrier layers, as well as on microscope pictures of the multilayers.

## 2. Multilayer design

Most materials in nature have a high absorption in the ~50–115 nm range, which makes them unusable for multilayer coatings because radiation cannot penetrate into the multilayer; hence, most coatings for this range are based on a single layer, which provides no filtering capacity. LiF is transparent above ~105 nm, which, in principle, makes it adequate for multilayers tuned above this limit, although to our knowledge such multilayers have not been reported. Below 105 nm, LiF becomes strongly absorbent, and it must be discarded. In and Sn are probably the only genuine low-absorbing materials in subranges within the 50–100 nm interval. In, with a ~60–100 nm low-absorption band, has a good transparency for multilayers in the present range, although its low melting point anticipates unstable multilayers, and hence it was also discarded.

Other than In, a source of low-absorption materials in the present range has been found among lanthanides, and many of them have been fully characterized recently (La [23], Tb [9], Gd [12], Nd [12], Yb [24,25], Ce [26], Pr [27], Eu [28], Tm [29], and Dy [30]), along with

materials with close chemical properties such as Sc [31–34]. This relatively low absorption makes them candidates for their use in novel multilayer coatings. Among these materials, Yb has one among the smallest absorption in the present range, and for this reason Yb was selected for the present multilayers.

The selection of the second material was performed in terms of a relatively low absorption and of a high contrast of the refractive index with respect to Yb. Al is a material with a relatively low extinction coefficient  $k$  for wavelengths close to or shorter than the Al plasma wavelength ( $\lambda_p = 83$  nm). Furthermore, Al refractive index  $n$  is much lower than unity and lower than the one of Yb in the range above  $\sim 70$  nm. However, both optical constants  $n$  and  $k$  of Al quickly vary with wavelength particularly close to the Al plasma wavelength, which makes difficult to figure out *a priori* the spectral performance of a multilayer based on Yb and Al. Furthermore, Al becomes increasingly absorptive and reflective above  $\lambda_p$ , and these properties suggest that multilayers based on Al may not work somewhat above  $\lambda_p$ .

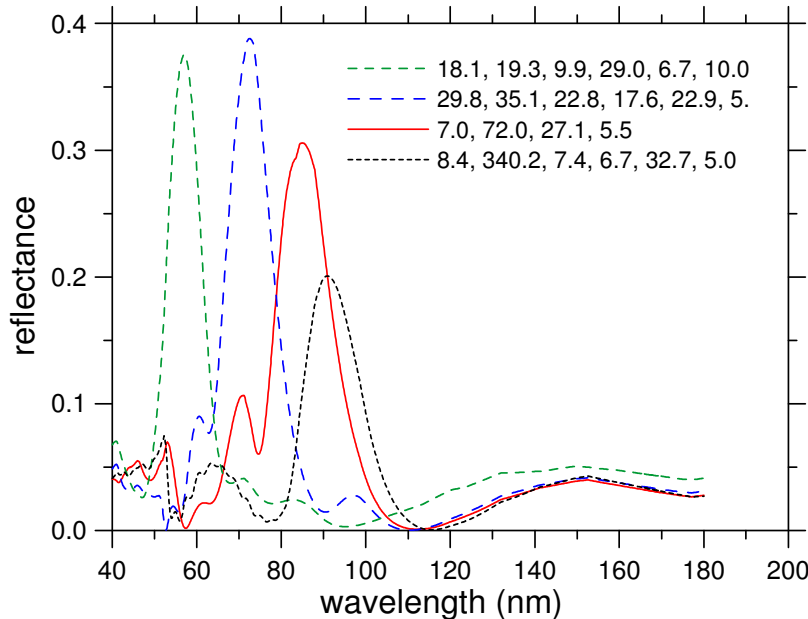


Fig. 1. Calculated reflectance versus wavelength for Yb/Al/Yb/SiO (peak at 85.8 nm) and Yb/Al/Yb/Al/Yb/SiO (peaks at 56.6, 73.5, and 90.9 nm) multilayers. Legend: film thicknesses in nm starting at the innermost layer.

One further difficulty is that bare films of Yb [24] and Al react with the atmosphere, which is expected to degrade the EUV performance of the multilayers. Therefore, we require the use of some protective coating on the Yb/Al multilayer. This problem does not have a straightforward solution since the ideal protective material would be transparent in the desired spectral range, but no such material is available and, in fact, we have selected Al and Yb for their relatively low absorption.

Let us see why we selected SiO for the protective layer of the multilayer. SiO is more transparent than SiO<sub>2</sub> in the spectral region from 50 to 113 nm [35]. Besides, one of the attractive properties of SiO is its relatively high vapor pressure; it evaporates at a much lower temperature than SiO<sub>2</sub> and condenses on cooler surfaces in uniform and adherent films. SiO is expected to be reasonably inert; Hass [36] studied the oxidation of the outermost layer of silicon monoxide films to silicon dioxide in air. The thickness of this layer depended on SiO deposition rate and on the residual vacuum pressure. In the best deposition conditions and at room temperature, Hass evaluated the thickness of the oxidized outer layer as  $\sim 5$  nm. Summarizing, SiO thin films are expected to be suitable as protective layers for the present

multilayers because of their relative inertness and low absorption, and of their good mechanical properties.

Multilayers were designed in the following way. We found that a simple Al/Yb/SiO multilayer (from the innermost to the outermost layer, one film per material) already displays interesting filtering properties in the EUV. In fact, the important absorption of Al above its  $\lambda_p$  impedes radiation to penetrate any further at these long wavelengths, which makes it unnecessary to add layers under Al for wavelengths above  $\lambda_p$ . However, the lower absorption of Al below  $\lambda_p$  enables radiation to penetrate deeper, so that the addition of further Yb and Al layers under the outermost Al layer can improve the performance in the short wavelengths.

Figure 1 displays examples of the calculated reflectance at normal incidence of various multilayer designs with peak wavelengths at 56.6, 73.5, 85.8, and 90.9 nm; multilayers with a reasonable peak at any wavelength between 50 and 92 nm could be designed. Multilayers were designed with one or two pairs of Yb/Al bilayers; multilayers were completed with an outermost Yb layer plus an SiO protective coating. Glass substrate was used in the calculations. Multilayers consist then in the sequence (starting with the innermost layer) Yb/Al/Yb/SiO (peak at 85.8 nm) and Yb/Al/Yb/Al/Yb/SiO (peaks at 56.6, 73.5, and 90.9 nm). In the calculations we used the optical constants available in the literature for Yb [24,25], Al [37–39] (Ref. 37 was used outside the ranges covered by Refs. 38 and 39), and SiO [35]. In the optimization, the layer thicknesses were allowed to vary, with the limit that the SiO protective coating was not thinner than 5 nm. A peak reflectance of 0.3 or larger could be obtained with bands centered at wavelengths at least between 50 and 88 nm. For Yb/Al multilayers with peaks above 88 nm, we found a fast peak-reflectance decrease, such as the multilayer of Fig. 1 with a reflectance of 0.20 at the peak of 90.9 nm. Calculations of Fig. 1 suggest that multilayers based on Yb and Al, with similar or larger reflectance than the reported in the literature (Refs [9–13]), are possible.

### 3. Experimental Techniques

The experimental system used in this work consists of a UHV thin-film deposition chamber connected in vacuum to a UHV reflectometer covering the 12–200 nm range. Freshly-prepared samples can be transferred to the reflectometry chamber without breaking vacuum. *In situ* transmittance and reflectance measurements can be performed at incidence angles going from  $3^\circ$  to  $\sim 87^\circ$  in two perpendicular planes of incidence. Ion and titanium sublimation pumps were used in both the deposition and the reflectometer chambers. After baking both chambers up to 470 K, their base pressure was  $\sim 2 \times 10^{-8}$  Pa. Liquid nitrogen in the Ti sublimation pumps was used to minimize pressure. The reflectometer-deposition system has been described with more detail elsewhere [40,41].

In the film deposition, we evaporated Al, Yb, and SiO of 99.999%, 99.9%, and 99.97% purities, using tungsten multi-stranded filaments, and Ta and Mo boats, at rates of 0.2–0.7, 0.07–0.13, and 0.03–0.07 nm/s, and under pressures of  $\sim 8 \times 10^{-6}$ ,  $\sim 1.7 \times 10^{-5}$ , and  $\sim 1.5 \times 10^{-7}$  Pa, respectively. The indications of Hass [36] regarding the adequate preparation of stoichiometric SiO thin films were followed. This requires the temperature of the evaporation source to be maintained at about 1520 K, with warmer (colder) sources giving rise to oxygen-depleted (oxygen-rich) films. The electrical parameters which provided a source temperature of  $\sim 1520$  K were calibrated with an optical pyrometer. Film thickness was monitored with a quartz crystal oscillator, which had been previously calibrated through Tolansky interferometry [42].

The films were deposited onto room-temperature float glass substrates. Reflectance measurements were performed both *in situ* on freshly deposited samples and after storage in a desiccator at room temperature. Reflectance measurements in the 200–800 nm range were performed *ex situ* at near-normal incidence by means of a Lambda 900 spectrophotometer using a URA accessory.

#### 4. Experimental results and discussion

Figure 2 shows the measured near-normal reflectance of two Yb/Al/Yb/SiO multilayers (starting at the innermost layer), with peak reflectance of 0.276-0.247 at 80-85 nm and a bandwidth of ~14.5-16 nm FWHM. Reflectance corresponds to samples not exposed to the atmosphere. Towards longer wavelengths, the multilayer passes a minimum reflectance close to 104.8-110 nm, and it increases longwards to values of 0.05-0.06. Towards shorter wavelengths, the sampling in one of the multilayers is too wide, and it does not resolve secondary peaks, which will be commented below.

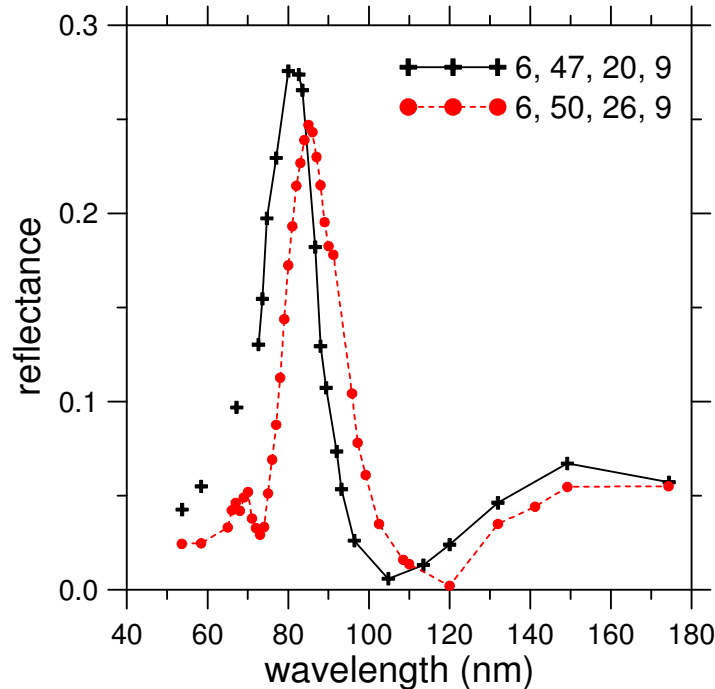


Fig. 2. Reflectance versus wavelength for two fresh Yb/Al/Yb/SiO multilayers. Legend: film thicknesses in nm starting at the innermost layer.

Figure 3 shows the reflectance measured in situ at three incidence angles ( $5^\circ$ ,  $30^\circ$ , and  $45^\circ$ ) for one of the multilayers plotted in Fig. 2. Measurements at  $30^\circ$  and  $45^\circ$  are the average of measurements at two perpendicular planes of incidence; these averaged values represent the reflectance that would be measured for non-polarized incoming radiation.

From Fig. 3 we observe that the band shifts towards shorter wavelengths when the angle moves away from normal incidence. The wavelengths at which the largest reflectance was measured were 80.0 ( $5^\circ$ ), 72.3 ( $30^\circ$ ), and 58.4 nm ( $45^\circ$ ). In order to know the wavelength at the reflectance peak with more accuracy, we calculated the thicknesses of the layers that best matched the reflectance measurements at the three incidence angles; the thicknesses are plotted in Fig. 2. In the calculations, we used the same optical constants referred above. The peaks were obtained at 79.3 ( $5^\circ$ ), 71.7 ( $30^\circ$ ), and 60.8 nm ( $45^\circ$ ). Between  $5^\circ$  and  $30^\circ$  the average shift approaches ~0.3-nm per degree. This peak shift with wavelength may be used as a means to spectrally tune multilayers.

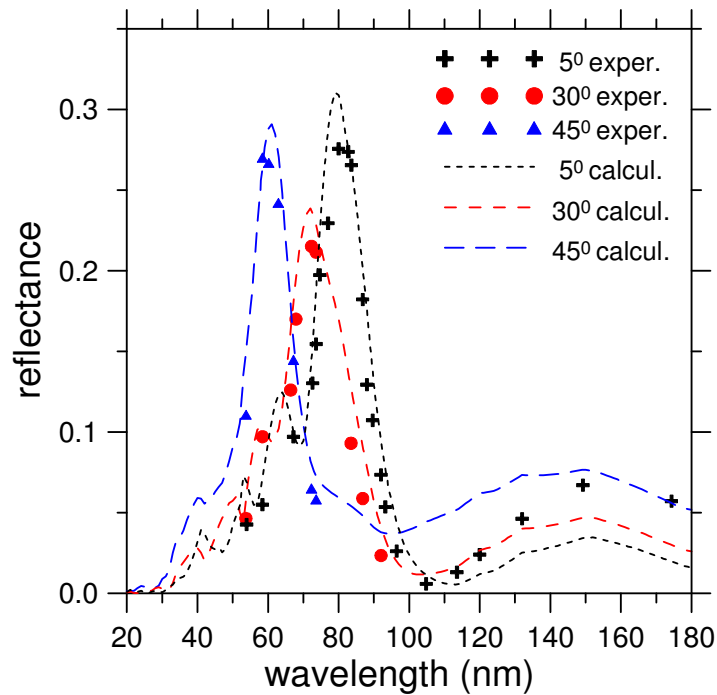


Fig. 3. Experimental and calculated reflectance versus wavelength for a fresh Yb/Al/Yb/SiO multilayer at three different angles of incidence

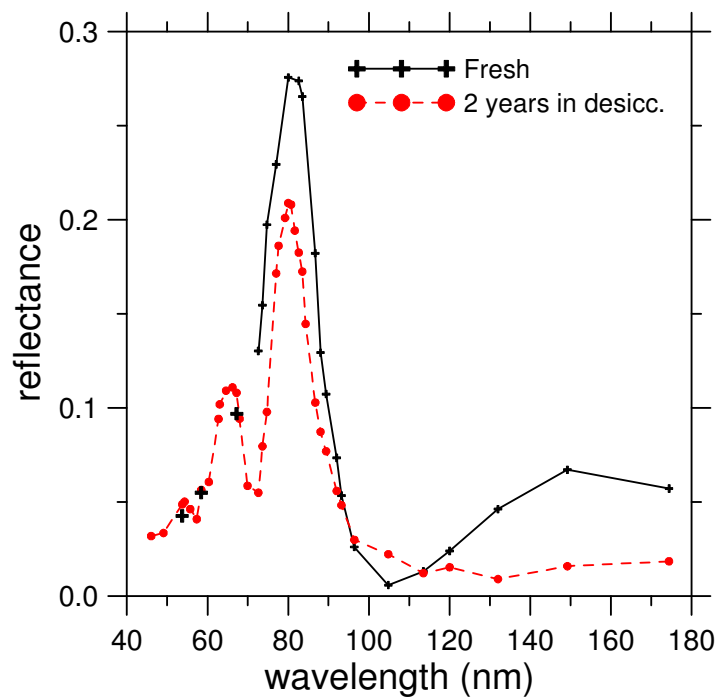


Fig. 4. Reflectance versus wavelength for an Yb/Al/Yb/SiO multilayer both fresh and after two years of storage in a desiccator

Figure 4 shows the reflectance of one of the multilayers plotted in Fig. 2 both fresh and after two years of storage in a desiccator. The reflectance peak at 80 nm has decreased to a

level of  $\sim 0.2$ . Hence, in spite of the decrease, the multilayer keeps a valuable efficiency over time. A finer sampling was used in the measurements of the aged multilayer, which displays the presence of a secondary peak that was not resolved when fresh. The reflectance decrease may arise from oxidation or contamination of the outer SiO film and/or from reactivity at the interfaces. The exact extent of the oxidation of the SiO film from monoxide to dioxide has been measured for a limited range of deposition parameters [36] and this extent is not known in general. In spite of that oxidation, the multilayers kept a valuable reflectance after a long exposure to the atmosphere, which confirms the efficiency of the 9-nm thick capping layer.

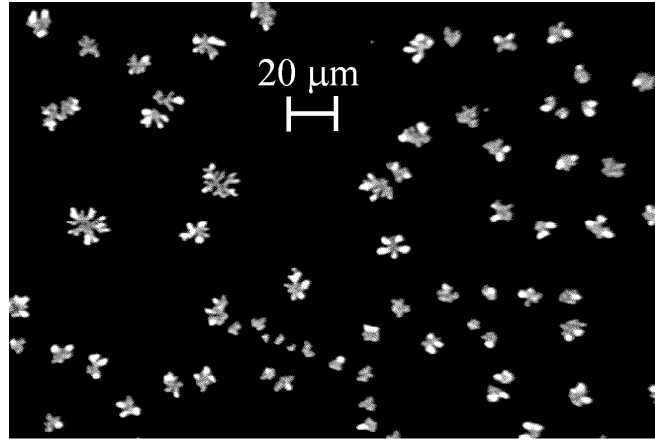


Fig. 5. Microscope view under transmitted light of an Yb/Al/Yb/SiO multilayer after one year of storage in a desiccator

The multilayers were observed under a microscope in transmitted light. Figure 5 displays a picture of a multilayer which shows that a dendritic structure has grown. The dendrites were observed immediately after extracting the samples from vacuum, and had a size of roughly  $2\ \mu\text{m}$ . Figure 5 corresponds to a multilayer stored in a desiccator for almost one year, and we could see that the dendrites grew over time.

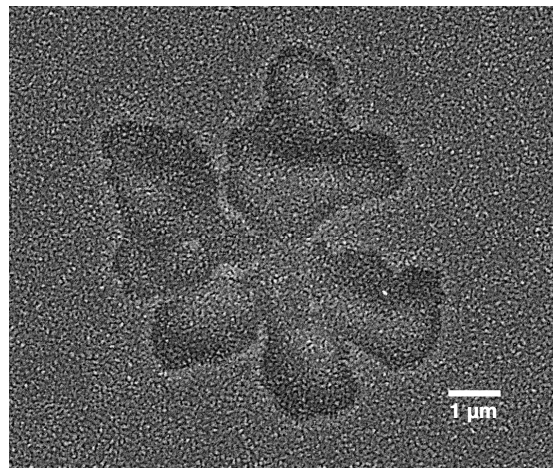


Fig. 6. SEM picture with a dendrite of an Yb/Al/Yb/SiO multilayer after two years of storage in a desiccator

Figure 6 displays an SEM picture of a multilayer after two years of storage in a desiccator. A dendrite can be observed with a good resolution. X-ray microanalysis was performed both at a dendrite and away from it. The chemical compositions were very similar for the different



materials (including the substrate) except for a large decrease in the abundance of Al at the dendrite. This shows that the dendrite is a locus away from which Al has migrated. This is compatible with the observations under the microscope in transmitted light, in which the dendrites were areas of lower optical density.

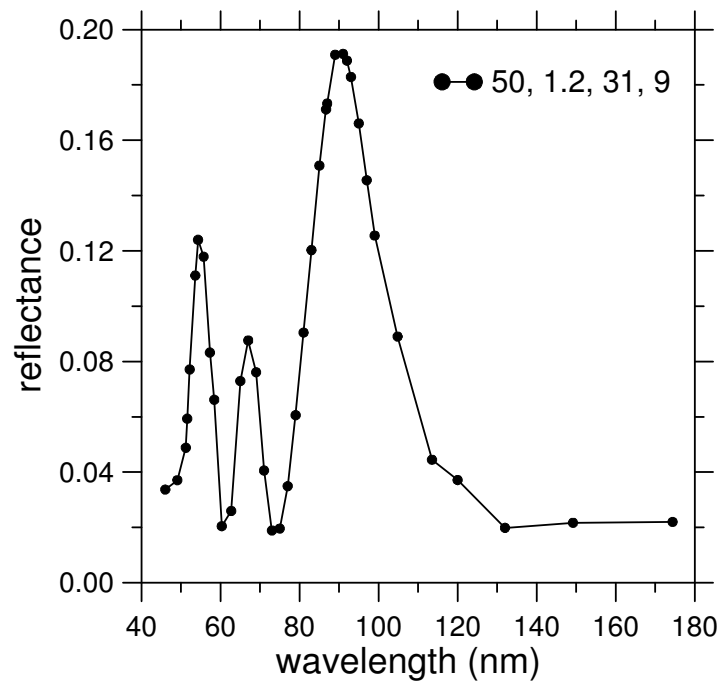


Fig. 7. Reflectance versus wavelength for an Al/SiO/Yb/SiO multilayer. Legend: film thicknesses in nm starting at the innermost layer.

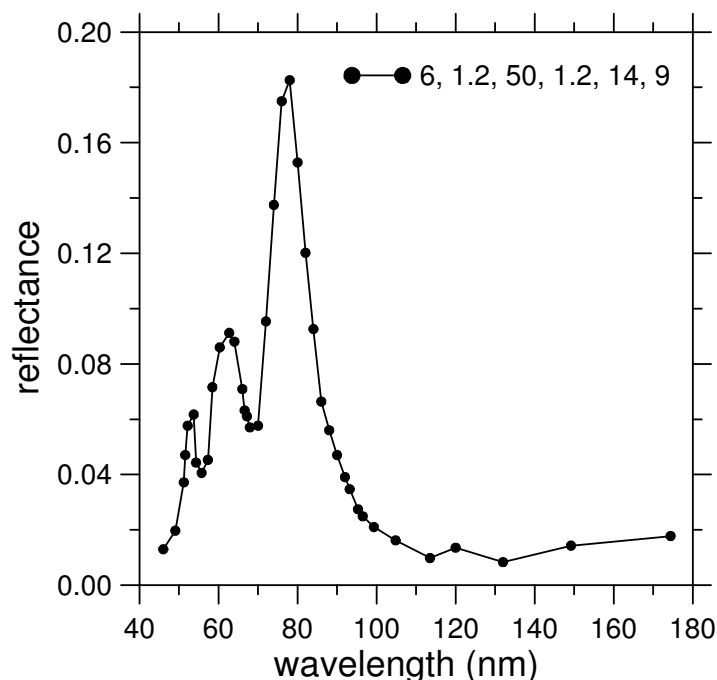


Fig. 8. Reflectance versus wavelength for an Yb/SiO/Al/SiO/Yb/SiO multilayer. Legend: film thicknesses in nm starting at the innermost layer.

The dendrite structure was then attributed to reactivity at the Yb-Al interfaces. In order to separate Al and Yb layers, new multilayers were prepared in which thin barrier layers of SiO were deposited at every interface between the two reactive materials. SiO was selected both for its relatively low absorption and because this same material had been successfully used for protective layers and it avoided the need for a fourth material.

Multilayers were prepared with barrier layers of various thicknesses; the depositions were performed in such a way that we deposited different barrier thicknesses on different areas of each sample. A 0.7-nm thick barrier layer was found not to avoid the dendritic growth or to avoid it only for a short time after deposition. Next attempted barrier thickness was 1.2 nm, which resulted in multilayers without any dendritic growth. Therefore, we decided to use 1.2-nm thick SiO barrier layers for the preparation of new multilayers.

Figure 7 shows the reflectance of an Al/SiO/Yb/SiO multilayer; it shows a peak reflectance of 0.191 at 91 nm and a bandwidth of ~22 nm FWHM. Measurements were performed on a sample that had been stored in a desiccator for 20 months. This relatively simple multilayer has a high peak reflectance at a remarkably long wavelength. However, it also presents two relatively high and undesired satellite bands, due to the simple multilayer design. These satellite bands can be significantly reduced with the addition of an inner Yb layer and even of further inner Al and Yb layers to the multilayer, as it can be seen in the calculated examples plotted in Fig. 1.

Figure 8 shows the reflectance of an Yb/SiO/Al/SiO/Yb/SiO multilayer. This multilayer shows a peak reflectance of 0.183 at 78 nm and a bandwidth of ~12 nm FWHM. Measurements were performed over a sample that had been stored in a desiccator for 20 months. This multilayer also presents satellite bands, but these bands are smaller compared to those of Fig. 7, which is due to the added innermost Yb layer. If we compare the data plotted in Figs. 4, 7, and 8, we find that the peak reflectance of aged multilayer both with and without barrier layers takes similar values of ~0.20.

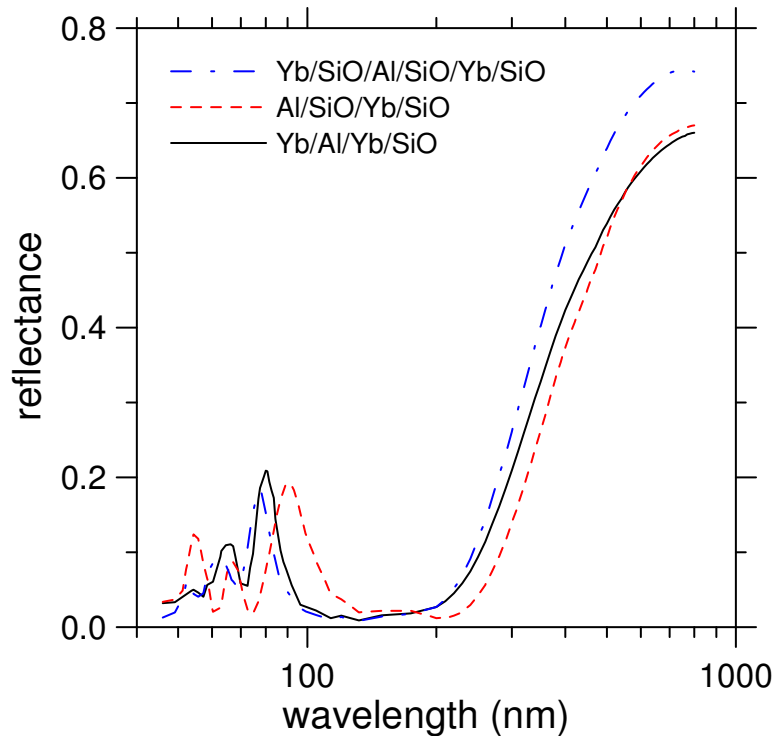


Fig. 9. Reflectance versus the logarithm of wavelength from the near infrared to the EUV for multilayers plotted in Figs. 4, 7, and 8

Multilayers with barrier layers were observed under a microscope after an ageing period of 20 to 24 months; no dendrite structure was observed on the aged multilayers.

Reflectance in the near UV, visible and near infrared was also measured. Figure 9 shows the reflectance in a broad spectral range for the aged multilayers displayed in Figs. 4, 7, and 8. The out-of-band reflectance is low below  $\sim 200$  nm. A reflectance lower than half of the main EUV peak is obtained at wavelengths lower than  $\sim 240$ – $280$  nm, whereas reflectance increases dramatically in the visible. The above rejection range is compatible with the use of these coatings in optical systems with detectors or cameras that are blind at long wavelengths. This is the case for micro channel plates (MCP) with photocathode materials such as CsI, KBr or KI, since they are blind to all wavelengths above their cutoffs, which are below 200 nm for all three materials.

The multilayers prepared in this research span a spectrum of peak wavelengths going from 78 to 91 nm. Designs enable predict that valuable multilayers can be prepared with peaks down to  $\sim 50$  nm, whereas a fast decrease in efficiency is predicted for peaks at  $\sim 92$  nm and above. If we compare with literature data, the multilayers prepared in this research are the only ones peaked above  $\sim 69$  nm, except for a  $B_4C/La$  multilayer of Ref [11], with a peak at 92 nm somewhat lower than the present multilayer at 91 nm; unfortunately, no data was reported for the  $B_4C/La$  multilayer above 100 nm and hence rejection efficiency cannot be compared. We plan further research to evaluate the stability of the coatings at increased temperature and to develop multilayer coatings with a larger number of layers in order to further minimize the size of the satellite bands shortward of the peak.

The present multilayers provide valuable performance covering a difficult spectral range. Yet, a narrow band cannot be efficiently covered with the present multilayers, mostly in the 92–115 nm range. In order to develop multilayers for the remaining gap, further research must be performed on the search of materials with relatively low absorption in the range.

## 5. Conclusions

A new type of narrowband filters based on Al, Yb, and SiO were designed and developed. The designs predict narrowband performance multilayers with peaks at least in the 50-92 nm range. Al and Yb were used for their relatively low absorption and high refractive index contrast; SiO films were used initially as protective layers, and finally also as barrier layers.

Yb/Al/Yb/SiO multilayers were prepared with valuable filtering performance. The different films of the multilayers were deposited by thermal evaporation in ultrahigh vacuum conditions. Multilayer near-normal reflectance was measured *in situ*, with a peak reflectance of 0.25–0.27 at wavelengths of 80-85 nm and a bandwidth of ~15-16 nm FWHM. The reflectance band shifted towards shorter wavelengths when the angle moved away from normal incidence; a shift of over 20 nm was measured from near-normal incidence to 45°. After a long period of storage in a desiccator, the peak reflectance retained a value of ~0.20.

Microscope observations revealed a dendritic structure, which was attributed to the reactivity between Al and Yb. X-ray microanalysis showed that the dendrite is a locus away from which Al has mostly migrated. A 1.2-nm thick SiO barrier layer between Al and Yb was found effective to avoid the formation of the dendrite structure. The reflectance of multilayers consisting of Yb/SiO/Al/SiO/Yb/SiO and Al/SiO/Yb/SiO showed narrowband performance with peaks in the 78-91 nm range. Multilayers with SiO barrier layers had grown no dendritic structure after ageing in a desiccator for ~2 years.

A low out-of-band reflectance was measured at wavelengths as long as ~200 nm for all types of multilayers prepared in this research, whereas the reflectance increased dramatically through the near UV towards the visible. This rejection range is compatible with the use of these coatings in optical systems with detectors or cameras that are blind at long wavelengths, such as detectors or cameras with photocathodes of CsI, KBr or KI.

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